

AMENDMENT UNDER 37 C.F.R. § 1.111  
U.S. Appln. No. 09/698,219

**REMARKS**

Claims 1-4 and 7-32 are pending in the present application. As will be discussed below, Claims 5 and 6 have been cancelled. No new matter has been added. Accordingly, entry of the present Amendment is requested.

Claims 5-7 have been rejected under 35 U.S.C. § 102(e) as being anticipated by U.S. Patent No. 5,958,532 to Krause.

Applicants respectfully submit, however, that since Claims 5 and 6 have been cancelled, the rejection has been overcome.

Claims 1-4 and 7 have been rejected as obvious over U.S. Patent No. 5,662,972 to Shimada *et al.* and the admitted prior art as modified by Shimada. Additionally, Claims 1-4, 8 and 11 have been rejected under 35 U.S.C. § 103(a) as being unpatentable over the "admitted prior art" in view of Shimada, Claim 9 has been rejected under 35 U.S.C. § 103(a) as being unpatentable over the "admitted prior art" and Shimada, further in view of Krause, and Claim 10 has been rejected under 35 U.S.C. § 103(a) as being unpatentable over the "admitted prior art" and Shamada, further in view of U.S. Patent No. 6,307,145 to Kataoka *et al.*

Applicants respectfully traverse these rejections for the following reasons.

The present invention is directed to a method of preparing a laminate having a fluorine resin and a crosslinked elastic adhesive body directly bonded to each other.

More particularly, the present invention provides for improved direct bonding between the resin and the crosslinking body, producing strongly bonded laminates having excellent properties such as storage stability, surface protection, stain resistance, high transparency and sealing.

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The improved bonding properties are achieved by treating the fluorine resin surface before lamination of the crosslinking elastic adhesive body thereto. Treatment of the surface is by one of the methods (1) to (3) described hereinbelow.

- (1) A method for the surface treatment of a fluorine resin wherein the surface of the resin has an absorbance at 360 nm of not smaller than 0.02/100 cm<sup>2</sup> when measured by iodometry.
- (2) A method for subjecting a fluorine resin surface to corona discharge treatment in an atmosphere of a nitrogen gas while controlling the concentration of an oxygen gas to 4 to 150 ppm.
- (3) A method for carrying out a thermal treatment after having subjected a fluorine resin surface to corona discharge treatment in air.

As described in the specification, treatment in accordance with the present invention increases the energy of the fluorine resin surface (Examples 1 and 2), producing acidic, reductive or radical-generating functional groups that enhance bonding between the resin and the crosslinking body upon crosslinking. Lamination of the crosslinking body to the surface-treated fluorine resin produces a very strongly bonded laminate with advantageous properties, such as storage stability as demonstrated in Examples 1 to 8 of the present specification.

Shimada discloses a process for producing a tubular fluorine-containing resin laminate which comprises, discharge-treating an outer surface of a tubular fluorine-containing resin molded article, extruding a molten resin onto the treated surface and coating the treated surface with the molten resin. Shimada discloses the treatment of fluorine resin by corona discharge treatment in an inert gas atmosphere containing an organic compound having a functional group.

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It is asserted, regarding Claims 1 and 2, that the person skilled in the art would have known that the reactivity of the fluorine resin after being subjected to a corona discharge treatment, is directly related to the intensity and duration of the treatment, and to determine these treatment conditions for optimum bonding would not have required undue experimentation.

However, Shimada fails to disclose a corona discharge treatment according to the present invention. In Shimada, the use of the organic compound having the functional group in the atmosphere of the discharge treatment is indispensable so as to introduce a functional group of an organic compound into the surface thereof or to graft-polymerize a polymerizable organic compound on the surface thereof. In contrast, the present claimed invention does not require such organic compounds nor graft-polymerization. The surface of the fluorine resin treated by the method of the present invention contains the radical generating functional groups so that the surface condition of the fluorine resin is quite different from the one obtained by Shimada's method.

The present inventors have found that a fluorine resin, which is treated so that it has the presently claimed absorbance at 360 nm, is excellent in bonding. The thus treated fluorine resin provides a laminate with a crosslinking elastic adhesive body which is integrally combined together. In this way, the fluorine resin and the crosslinking elastic adhesive body are bonded strongly, thereby making it possible to reliably make a laminate that has high storage stability, surface protection, stain resistance, high transparency and a member-sealing property.

Referring to page 9 of the specification, it is indicated that the specification requires a corona discharge treatment under routine conditions, and that there are no examples of a surface treated fluorine resin having absorbance values less than the claimed range.

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However, as discussed above, there is a fundamental difference between the inventive method and Shimada's method. In particular, there would not be obtained the radical-generating function groups on the surface of the fluorine resin by Shimada's method which would result in almost zero absorbance at 360 nm. Accordingly, those skilled in the art would not have known that the iodometric measurement gives the optimum bonding condition as disclosed in the present invention. Further, Examples 1, 2 and Comparative Example 1 show the energy density and absorbance at 360 nm. Even though the surface-treated fluorine resins had different quantities of energy density (Original Example 1 had half the energy density of Original Example 2), that is, the conditions of the treatment such as the intensity and duration varied, in so far as the absorbance at 360 nm is 0.02/100 cm<sup>2</sup> or more, the desired bonding force can be obtained. This leads to a laminate in which the fluorine resin and the crosslinking elastic adhesive are strongly bonded each other. Original Examples 1 and 2 demonstrated almost the same high bonding force regardless of the difference of energy density.

Thus, in accordance with the present invention, it is quite important to introduce radical generating functional groups to the surface of the fluorine resin, so that the laminate having a satisfactory bonding to the crosslinking elastic adhesive body can be obtained. To achieve a laminate having this excellent property, iodometric measurement is employed to determine the quantity of radical generating functional groups on the fluorine resin surface. If the absorbance is less than 0.02/100 cm<sup>2</sup>, the amount of the radical generating functional groups is too small to impart satisfactory bonding to a crosslinking elastic adhesive body, which fails to ensure direct and strong bonding to a crosslinking elastic adhesive body. Thus, Shimada and the admitted prior art fail to disclose the present claimed invention.

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Accordingly, Applicants submit that the cited references fail to subject a fluorine resin to surface treatment so that an absorbance at 360 nm is 0.02/100 cm<sup>2</sup> or more when determined by iodometry. Thus, Applicants respectfully submit that the present claimed invention is patentable over Shimada and the admitted prior art modified by Shimada.

Regarding Claims 3 and 4, it is asserted that the person skilled in the art would have readily appreciated that the atmosphere would contain some negligible oxygen concentration or a trace amount of oxygen.

However, as mentioned above, Shimada is directed to a corona discharge treatment in an inert gas atmosphere containing an organic compound having a functional group, not treatment in an inert atmosphere with the presently claimed amount of oxygen content. Surface treatment in an atmosphere of nitrogen with 4 to 150 ppm oxygen in accordance with the present claimed invention produces a laminate in which the fluorine resin and crosslinking elastic adhesive are strongly bonded together with superior storage stability, and minimized deterioration in the bonding force after storage for one month. Original Examples 3 to 7 and Comparative Examples 2 to 4 show that acceptable storage stabilities of 80% or above were only observed for oxygen gas concentrations in the range 4 to 150 ppm (page 28, Table 2). Departure from this range (from 134 to 460 ppm) resulted in a relatively large drop (12 percentage points) in the storage stability of the laminate (Original Example 7 and Comparative Example 3). Plus, when the oxygen gas concentration exceeds the optimum range, (Comparative Example 4), initial bonding force is considerably lowers and what is worse, bonding force after one month and storage stability could not be evaluated and the thus obtained laminate was completely out of the question.

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As described above, controlling the concentration of oxygen gas to within the range of 4 to 150 ppm in the nitrogen gas is quite important when subjecting corona discharge treatment to the fluorine resin in order to achieve the drastically improved bonding property between the fluorine resin and the crosslinking elastic adhesive body and storage stability of the laminate. Shimada is not directed to improving the storage stability of the laminate as disclosed in the present invention. Even though a person skilled in the art would have known that there exist a trace amount of oxygen in the atmosphere of the discharge treatment as the Examiner alleges, Applicants respectfully submit that it could not have been obvious from the cited references that the optimum range of oxygen concentrations as defined in the present claims imparts the excellent laminate storage stability (minimized deterioration in the bonding force).

Accordingly, Applicants respectfully submit that surface treatment of a fluorine resin, comprising subjecting the surface of a fluorine resin to corona discharge treatment in a nitrogen gas atmosphere while controlling a concentration of an oxygen gas within a range of 4 to 150 ppm is not expected from Shimada and the admitted prior art.

Regarding the remaining references, Krause discloses a fluoropolymer composite tube formed by a process comprising the steps of forming a fluoropolymer substrate and layering it with a thermosetting or thermoplastic elastomer. Krause teaches that the fluoropolymer such as ethylene-tetrafluoroethylene (ETEE) is extruded by melt extrusion, on which thermosetting or thermoplastic elastomer material is extruded and cured. The surface of the fluoropolymer is treated (prior to extruding elastomer) by plasma discharge or corona discharge.

Unlike in the present invention, in Krause, heat treatment for curing the thermosetting elastomer is effected after the fluoropolymer is combined with the elastomer. In contrast, in the

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present invention, thermal treatment is effected to the fluoropolymer before lamination of the crosslinking elastomer, thereby the laminate having a dramatically improved bonding force is obtained as demonstrated in Original Example 8 and Comparative Example 5.

Krause fails to disclose the present invention of subjecting the fluorine resin surface to corona discharge in air with subsequent thermal treatment prior to laminating the crosslinking elastic adhesive body thereto, as recited in Claim 8. Even if Krause discloses the use of the crosslinking elastic adhesive body, the present invention is not obvious over Krause.

Furthermore, Krause uses an amine curing agents, such as triethylene tetramine, along with the thermosetting materials (see, lines 59-65 in column 6). The present invention does not use amine curing agents. An organic peroxide may be used for thermal curing and/or as radical photopolymerization initiator for photopolymerization, together with ethylene-vinyl acetate copolymer. Thus, the laminate comprising fluorine resin and ethylene-vinyl acetate copolymer according to the present invention has excellent storage stability as well as improved bonding force.

Kataoka relates to a solar cell module in which a surface of a light incident side of a photovoltaic element is sealed by a covering member comprised of at least two layers including a sealant resin layer of a transparent, organic polymer resin and an outermost, transparent surface protecting film. Kataoka teaches that the transparent surface sealant resin layer contains a crosslinked resin such as ethylene-vinyl acetate copolymer resin crosslinked by an organic peroxide.

However, there is no disclosure of the methods of the present invention, and those skilled in the art would not be motivated to combine the references to arrive at the present invention.

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In view of the above, reconsideration and allowance of this application are now believed to be in order, and such actions are hereby solicited. If any points remain in issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the telephone number listed below.

The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

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PATENT TRADEMARK OFFICE

Date: July 1, 2003

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**APPENDIX**  
**VERSION WITH MARKINGS TO SHOW CHANGES MADE**

**IN THE CLAIMS:**

Claims 5 and 6 are canceled.